Laser structuration of dielectric materials by a train of femtosecond pulses through cumulative effects

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1. Introduction and general phenomenology of the physical processes

Optical materials can be structured by laser pulses to get new material functionalities in various scientific area going from photonics to medicine. For instance, wave guides, nano-gratings, emergence of nonlinear optical properties for data storage [1], cutting and welding of materials are applications of great interest. Structuration driven by a train of laser pulses is strongly emerging due to its advantages: table top laser facility, very well controlled structuration with energy deposition accuracy in the nJ range by adjusting the number of pulses, etc. The material structuration due to pulse-to-pulse cumulative effects should be deeply understood to design specific structures. This may be achieved by modelling the main physical processes and their possible coupling. Briefly, each laser pulse first induces photo-ionization and heats the conduction electrons which can then transfer their energy to the lattice. That leads to a local increase in the material temperature together with heat diffusion and thermally-activated ions migration on longer timescales. Since the laser pulse is partially absorbed, the electron dynamics and the pulse propagation are closely coupled. Due to the low heat diffusion coefficient of dielectric materials, the laser energy may be accumulated in the absorption region, leading to high temperatures even if the single pulse energy is too low to induce itself any significant material modification. A general modelling including all the above-mentioned processes will be presented, including the two following applications of interest.

2. Nanostructures formation in silver-doped phosphate glasses

Here a phosphate glass doped with initially uniformly distributed Ag^+ ions is irradiated. The laser-induced ionization first leads to the formation of Ag_0 atoms through the reaction $Ag^+ + e^- \rightarrow Ag^0$. The latter specie may then associates with Ag^+ to form Ag_2^+, which exhibits luminescence properties rendering it possible to observe their spatial distribution. Fig. 1(a) shows the emergence of a micrometric ring structure after an irradiation by 10^7 laser pulses [1] with parameters: 1030 nm, 470 fs, 1.2 μm of waist, 100 nJ, 10MHz. Due to the migration of charged species, a static electric field originates leading to the formation of local nonlinear optical properties. A model based on laser heating, heat diffusion, and thermally-activated diffusion and kinetic reactions of the various silver species, allows us to account for the observed structure as shown in Fig. 1(b). The details of the modelling, mechanisms for cumulative effects, and influence of laser parameters on the ring characteristics will be presented.

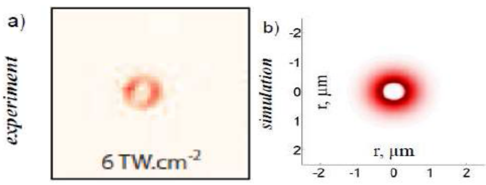

Fig. 1. (a) Experimental and (b) predicted ring structure (Section 2).

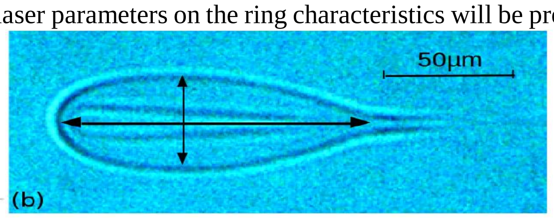

Fig. 2. Laser affected zone in sodalime (Section 3)

3. Refractive index modifications in sodalime glasses

Sodalime glasses irradiated by a train of 500 laser pulses with various repetition rates (RR) is addressed. Each pulse with 300fs duration, 1030nm wavelength and micro-joule energy, is focused by a 10x objective in the glass. As shown by Fig. 2, the material affected zone exhibits a particular comet-like shape with characteristic longitudinal and transverse lengths. Experimental investigations show that this structure only appears for RR larger than roughly 100kHz. Then, both lengths increase with respect to the RR up to 500kHz. In order to shed light on the underlying cumulative effects taking place, the energy deposition is obtained by simulating the laser pulse propagation through a Forward Maxwell code [2]. The thermal effects are modelled by solving a heat diffusion equation with the deposited laser energy as initial condition. The softening temperature is considered as the threshold for a permanent index modification of the matter. The obtained evolutions of both characteristic lengths with respect to the RR are in a good agreement with the experimental data, demonstrating that the material modifications originate from heat accumulation and subsequent phase transition.

[1] G. Papon *et al*, "Fluorescence and second harmonic generation correlative microscopy to probe space charge separation and silver cluster stabilization during direct laser writing in a tailored silver-containing glass," Optical Materials Express **3**(11), 1855-1861 (2013).

[2] L. Bergé *et al*, "Ultrashort filaments of light in weakly ionized optically transparent media," Rep. Prog. Phys. **70**, 1633 (2007).